Manipulating Quantum Systems with Ultrashort Laser Pulses

Suxing Hu and Lee Collins (T-4);
suxing@lanl.gov

Lasers producing ultrashort pulses have issued in a new era of investigation into the interplay of radiation with matter. Such pulses, now shorter than a few femtoseconds, can produce very intense electromagnetic fields, exceeding the basic binding of the electrons to atoms and molecules. Such fields have initiated a variety of phenomena such as Coulomb explosions, multiphoton ionization, and high harmonic generation. On the other hand, pulses in the short-time mode at low intensities permit the manipulating and probing of very intricate atomic, molecular, and condensed matter processes. Examples of such processes include tracking the evolution of a vibrational wave packet or changing the outcome of a collision. Few-cycle pulses (FCPs) provide one particularly useful form that allows illumination of the target for a very brief period with a well-characterized field, effectively constructed to drive a particular process. We have investigated several applications of these ultrashort pulses, including high harmonic generation [1], redistribution of Rydberg populations [2], recombination [3, 4], and molecular imaging [5].

Molecular Imaging: The use of electron diffraction to probe the structure of materials has a long history. The basic experiments involve bombarding a target with an external electron beam and observing the resulting diffraction pattern. With ultrashort pulses, on the timescale of molecular processes, a situation exists for using the molecule's own electron to effect the scattering. A FCP is constructed so that the initial part ionizes a valance molecular electron. Once the electron has reached a sufficient distance from the target, the field is reversed, and the electron is driven back through the molecular core, scattering from the constituent particles. The diffraction pattern from this electron can then be observed at some large distance. Figure 1 shows the basic process for a FCP laser field interacting with a K_2+ molecule. We use a molecule consisting of heavy atoms so that the internuclear separation remains relatively fixed during the interaction due to the long vibrational period. We have investigated this process using large-scale computer simulations that solve the 3D time-dependent Schrödinger equation on spatial and temporal grids. In Fig. 2, we display the characteristic diffraction pattern for the electron that results from the FCP interaction. Even more encouraging, from the analysis of this pattern, we can extract information on the internuclear separation. Experiments along this line are currently underway in Canada.

High Harmonic Generation: A very intense laser field impinging on an atom causes the atom to radiate in very narrow peaks

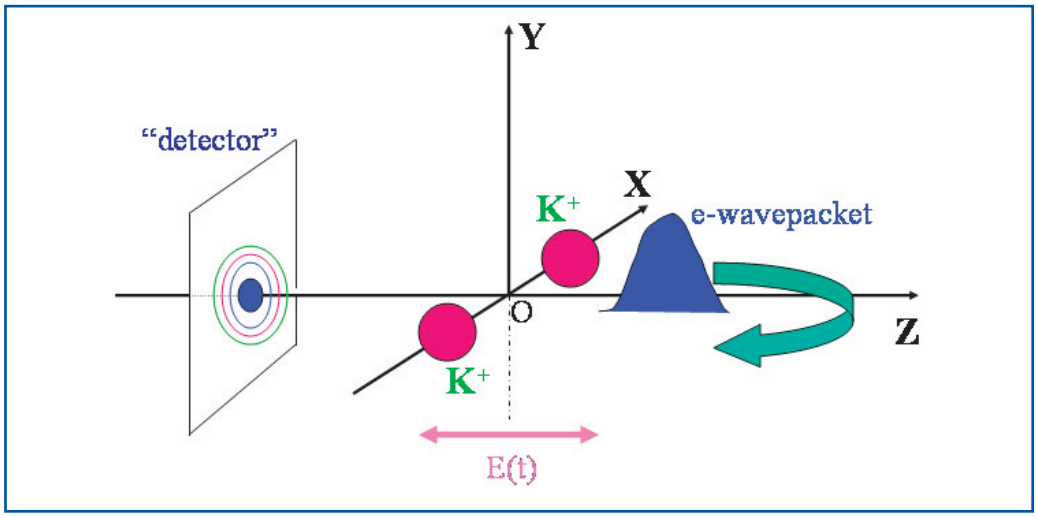

Figure 1— Schematic of the diffraction of a molecular electron ionized and then driven back through the target by a short-pulsed laser field.

at harmonics of the laser frequency. The intensity in the high harmonics drops by many orders of magnitude from that of the driving field, but, since the initial intensity was so large [> 10^{15} W/cm^2], these harmonics emerge at strengths both observable and usable for other applications. For neutrals, the limit in the practical harmonic spectrum comes at about 500 eV. The use of ions could push this limit much higher into the X-ray regime; however, the density of trapped ions remains very low compared to neutrals as does the photon output. We have explored a hybrid mechanism that employs a Rydberg atom in an intense field. Neutral Rydbergs can be produced in far larger quantities than ions. The outer loosely bound electron is stabilized against ionization by the intense field so that the system remains basically neutral. On the other hand, the field also interacts with the tightly bound ionic core, generating harmonics much as with ions. Figure 3 shows the harmonic generation for this system with a field intensity of 1.2 x 10^{16} W/cm^2 and a photon energy of 2.36 eV. Since reasonable intense harmonics up to the order of 500 are produced, this means the system generates moderately intense, very short-pulsed X-rays (~ k eV). Such sources of ultrashort, coherent X-rays have a large variety of applications in biology, chemistry, and plasma science.

[1] S.X. Hu and L.A. Collins, *Phys. Rev. A* **69**, 033405 (2004).
[2] S.X. Hu and L.A. Collins, *Phys. Rev. A* **69** 041402 (2004) [Rapid Comm.].
[3] S.X. Hu and L.A. Collins, *Phys. Rev. A* **70**, 013407 (2004).
[4] S.X. Hu and L.A. Collins, *Phys. Rev. A* **70**, 035401 (2004).
[5] S.X. Hu and L.A. Collins, *Phys. Rev. Lett.* **94**, 073004 (2005) (in press).

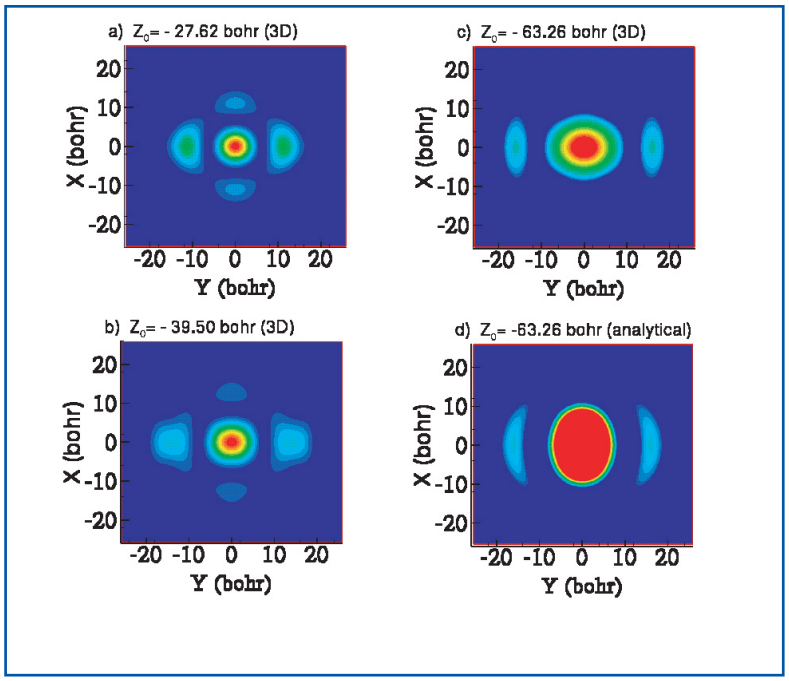

***Figure 2—** Diffraction patterns at various distances from the target of an electron driven through a molecule by a FCP. The "analytical" figure arises from a simple two-center independent scattering model.*

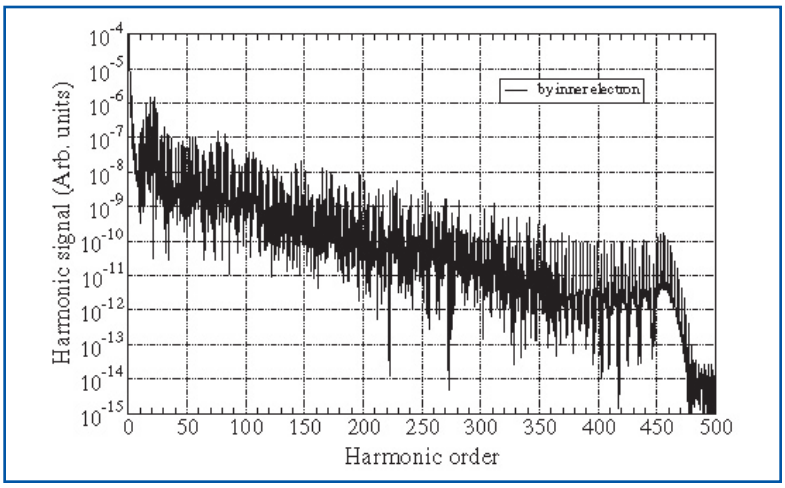

***Figure 3—** Harmonic spectrum generated from the inner electron of a Lithium Rydberg atom exposed to an intense laser field.*